Sorption Mechanisms for Mercury Capture in Warm Post-Gasification Gas Clean-Up Systems

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OBJECTIVES

This research is directed towards a sorbent injection/particle removal process, where sorbent may be injected upstream of the warm gas cleanup system to scavenge Hg and other trace metals, and removed (with the metals) within the warm gas cleanup process. The specific objectives of this project are to understand and quantify, through fundamentally based models, mechanisms of interaction between mercury vapor compounds and novel paper waste derived (kaolinite + calcium based) sorbents (PWDS), with and without substrate modifiers. Two types of interactions will be investigated:

1. *hot injection/warm removal*, i.e interactions with *original* PWDS at temperatures *higher* than 700˚F which allow the contaminants to be retained until the gases are cooled to warm gas clean up conditions where the spent sorbent is removed.

- *warm injection/warm removal* interactions with *modified* PWDS which allows sorption at temperatures *below* 700˚F and *simultaneous* warm gas cleaning at lower temperatures**.**

APPROACH

•Experimental flow reactor studies to investigate Hg and trace metal capture
  •Heated quartz reactor,Simulated gasification off-gases
  •Sorbent screening- PWDS, component mixtures, PWDS+Ca(ClO)$_2$
  •Mechanism elucidation

•Theory
  •Bonding mechanisms (computational, *ab-initio* calculations using Acelrys DMOL3 software
  •Extrapolation to high pressures and gasification conditions

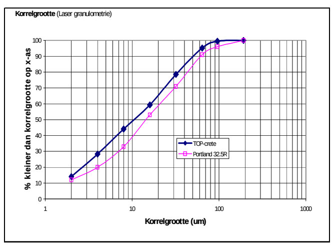
PWDS (Paper Waste Derived Sorbent) COMPOSITION

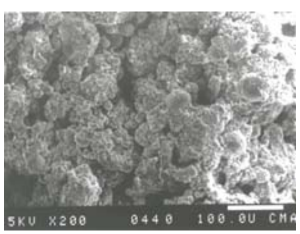
(for high/medium temperature Hg sorption)

Limestone	**$CaCO_3$**	**41%**
Metakaoline	**Al_2O_3. 2 SiO_2**	**29%**
Lime	**CaO**	**23%**
Inert		**7%**

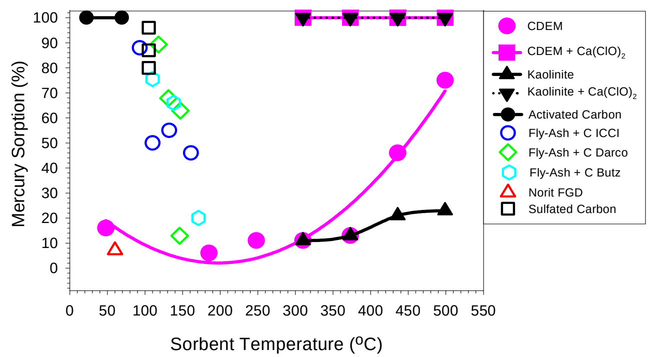
Trace components

MgO	2%
TiO_2	1%
Fe_2O_3	0.7%
SO_3	0.6%
K_2O	0.4%
P_2O_5	0.5%
CuO	0.1%
Na_2O	< 0.1%
BaO	< 0.1%
Cl	< 0.1%
MnO	< 0.1%
ZrO_2	< 0.1%
ZnO	0.03%
NiO	0.002%
PbO	0.005%
Cr_2O_3	0.002%
As_2O_3	< 0.001%
HgO 0.0001%	<
CdO	0.00005%

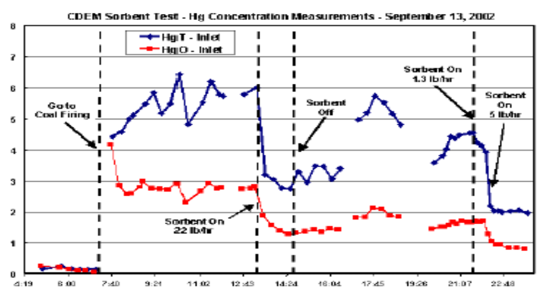




EXPERIMENTAL RESULTS

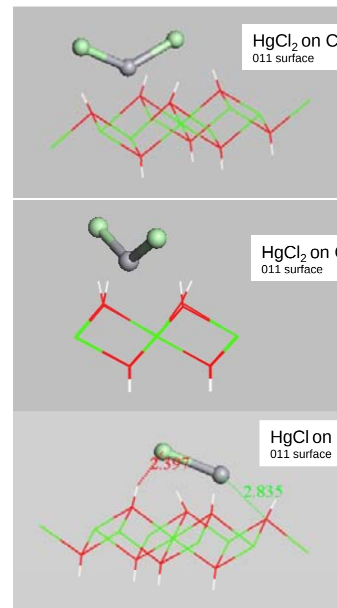


Packed bed results (shaded symbols, lines). Chemisorption data compared to literature physisorption data. Modified (with calcium hypochlorite) sorbent shows 100% capture at lower temperatures than original PWDS, but still higher than activated carbon and the like.



PWDS at/SRI, 1.75MW, semi-industrial scale test data. Total Hg (blue) and Hg, metal (red) trace in µg/m3. Choctaw coal. Sample before baghouse. In-flight capture at 13h-14:30h and at low sorbent injection at 22h.

COMPUTATIONAL RESULTS (ACELRYS DMOL3 Software)



Oxygen is represented by the red color while calcium is green and hydrogen is white. We see that mercury always binds most closely to the calcium instead of the oxygen atoms while the chlorine atoms are bent away from the surface despite there being some attractive force to the hydrogen on the surface.

This test will be ¼ inch tall (18 point) Only use the the very smallest text

Adsorption Species	Hg	HgCl	$HgCl_2$
Hg-Ca distance	3.002	2.835	2.606
Cl-H distance	N/A	2.397	2.073/2.330
Adsorption energy (kcal/mol)	-15.69	-40.29	-43.68

As the mercury is oxidized it binds much more strongly to the surface. This is shown by both the increase in binding energy and the shortening of the bonds to the surface atoms**.**